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The Effects of Isotopic Separation on Closed Nuclear Fuel Cycles

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The Effects of Isotopic Separation on Closed Nuclear Fuel Cycles

by

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Dedication

I'd like to dedicate this work to my parents, Hugh Flanagan and Patricia Korta, for their love and support on my journey up to this point. Throughout my entire academic career they have stood by me and my decision to further my education.

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Abstract

The Effects of Isotopic Separation on Closed Nuclear Fuel Cycles

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The University of Texas at Austin, 2012

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This paper investigates the potential benefits to the fuel cycle outcomes of removing a single isotope during separation processes. Two strategies for managing the removed isotope are considered. The first strategy looks at removal of a short to intermediate lived isotope from a mass stream to be recycled and subsequently recycling its decay daughter in a transmuting reactor. The second investigates the effect of removing a long lived fission product from high level waste and recycling it into the transmuting reactor. This analysis shows that the removal of Cm-244 using the first strategy provides a marked benefit to several fuel cycle metrics. The second strategy benefits the long term radioactivity measured from the high level waste from isotopes including Zr-93 and Cs-137.

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1.0 Introduction

The role of separations processes in advanced fuel cycles has historically been limited to partitioning technologies that operate at the elemental level. These processes exploit the chemical properties of the different transuranic material and fission products to be separated from a nuclear fuel cycle mass stream. A reprocessing technology known as PUREX separates plutonium from the rest of the actinides present in spent fuel. The UREX process will separate uranium from other major actinides. Other processes are capable of separating out elemental streams of individual actinides or fission products, such as technetium and iodine [1].

These reprocessing techniques are effective for dealing with plutonium to be burned in a fast spectrum reactor. For some other actinides exposure to a fast spectrum leads to neutron capture reactions. Through these capture reactions and the decays that follow high order actinides are produced. These higher order actinides would otherwise not be produced if their parents were not present in a fast spectrum reactor. An isotope that leads to the products of high order actinides in this manner is termed a 'gateway isotope'. A good example of a gateway isotope is Cm-244. The presence of Cm-244 can lead to the creation of many isotopes that are more difficult to burn or handle such as Cm-247, Cm-248 and isotopes of berkelium and californium. It takes three species neutron-gamma capture reactions to produce Cm-247 from Cm-244. These species are

created through

$$Cm_{96}^{244} + n_0^1 = Cm_{96}^{245} + \gamma$$

$$Cm_{96}^{245} + n_0^1 = Cm_{96}^{246} + \gamma$$

$$Cm_{96}^{246} + n_0^1 = Cm_{96}^{247} + \gamma$$

Californium and berkelium are produced from further neutron captures and subsequent beta decays. Californium , Cm, and Bk isotopes often have decay modes of spontaneous fission. Californium 252, for example, has a spontaneous fission branching ratio of 3.09%. Spontaneous fission makes handling of spent fuel more difficult due to the difficulties associated with shielding against a neutron radiation field. This makes the fabrication of fast spectrum reactor fuel more expensive. Reducing the neutron field would therefore provide a direct benefit to the fuel cycle.

In addition, isotopic separation and transmutation prior to disposal may be desirable for isotopes whose waste impacts would be reduced if transmuted. This is the case for the long lived fission products. These fission products will remain radioactive in a geological repository for millions of years. In such cases elemental separation may not be feasible. Other isotopes of the element may be the neutron capture parent of a long lived fission product and therefore produce more of long lived material than this process aims to destroy.

Cesium has only one isotope of interest to be transmuted, Cs-135. Cs-135 has a half-life of 2.3 million years. Through neutron capture it becomes Cs-136 which has a half-life of only 13.16 days, and decays to stable Ba-136. This is the desired reaction for dealing with Cs-135 as Ba-136 does not have storage difficulties.

The addition of other elements of cesium complicates matters. Cs-134 decays to stable isotopes with half-life of 2.06 years respectively. Cs-133 is stable. All of these isotopes are present in spent fuel from nuclear reactors. Chemical separation alone would mean that these isotopes would also be subjected to the neutron flux being used to transmute Cs-135. Neutron capture reactions could transmute any of these into heavier cesium atoms. For Cs-133 this would mean transmuting a stable nuclide into Cs-134 and ultimately Cs-135, producing the isotope that is meant to be removed. Additionally burning of chemically separated cesium would introduce Cs-137. Cs-137 is a short lived fission product and therefore handling of this material would be very costly and dangerous.

The ability to partition specific isotopes could therefore augment the benefits to fuel cycle outcomes that can be achieved through of chemical separation alone. Indeed, fuel cycle strategies involving partitioning of specific isotopes, both actinides and long lived fission products, have been considered [2][3]. These strategies often look at only a single isotope and a single method for disposal. In some of the studies the isotope is not partitioned from the elemental mass stream it belongs to. Instead the whole mass stream

is treated in an attempt to transmute the problem isotope. In addition to giving rise to undesirable transmutations mentioned above, this increases the amount of mass being passed to fuel cycle facilities that could otherwise be smaller if only the isotope in question were being investigated.

This study is predicated upon the development of a cost-effective isotopic separation technique suitable for elemental streams containing a number of radioisotopes. Many candidates, most of which rely upon selective laser excitation of a chemical or nuclear state, are described in the literature [4]. In one promising technique [5], the material in elemental form is vaporized in a high temperature oven and entrained in a high velocity stream of inert gas. [5].

The high velocity beam is then passed through an array of lasers used to optically pump the selected isotope into an excited state. The modified electron structure creates a modified magnetic moment for the atom. This change is exploited using a magnetic field to divert the excited isotopes into a separation collection chamber.

The process has a theoretical enrichment capability for lithium-6 of 95% in the enriched stream. After one pass 63.2% of the lithium-6 was removed from the waste stream. The amount of material passed through this technique is low, but only isotopes with small weight fractions within the fuel cycle are considered for this technique. Large components, like uranium, are not separable with this technique.

Increased separation efficiencies are obtained by constructing a cascade of separation devices. The level of separation or enrichment depends only on the number of stages the material is passed through and is therefore easily controlled. For this study the total enrichment is assumed to be 100% such that pure product streams are produced.

The aims of this work are to quantify the possible benefits of isotopic separation in the nuclear fuel cycle and to derive an approximate design target cost below which the isotopic separation technology must fall if it is to be attractive.

This investigation uses an open source fuel cycle simulator developed at the University of Texas at Austin called Bright [6]. Bright is a fluence based reactor model that accepts input fuel isotopic vectors and returns a spent fuel isotopic vector. Output isotopic vectors are then passed to Origen 2.2 [7] to computer the metrics. Origen 2.2 is also used to decay the spent fuel material out to one million years.

A collection of metrics will be used to determine the impact on cost. The metrics describe important characteristics of the nuclear fuel cycle including radioactivity, cumulative decay heat and spontaneous fission activity. Radioactivity and spontaneous neutron source are measured at various points during the existence of spent nuclear fuel depending on when these values are of importance.

This work compares a base case fuel cycle to perturbed versions that incorporate

isotopic separation. The base case fuel cycle is a nuclear fleet composed of light water reactors and fast spectrum reactors. The metrics will be calculated for this standard case. Perturbations will then be applied to this cycle to investigate the impacts of various methods of isotopic separation. In the first case an actinide isotope is removed from spent fuel and allowed to decay for a set number of half-lives. The daughter products are returned to the fuel cycle for recycling. An example of this first perturbation is removal of Cm-244. Cm-244 is allowed to decay to Pu-240, which is then incorporated into the fact reactor fuel to be transmuted. In the second perturbation long lived fission products are removed from spent fuel and recycled inside a fast reactor or inside a linear accelerator. In this case examples of long lived fission products to be treated are Cs-135, I-129 and Zr-93.

This work is done to establish economically viable strategies for dealing with problem isotopes within the nuclear fuel cycle. Past studies of isotopic separation applied to the nuclear fuel cycle have stopped short of addressing the economic viability of the separation technology. This work will establish cost targets and quantify benefits of isotopic separation to the nuclear fuel cycle.

This thesis is structured as follows. Section 2.0 reviews and summarizes important work used in this investigation. The method used to analyze the fuel cycles is discussed in section 3.0. Results of the investigation are reported in section 4.0 with conclusions based on these results found in 5.0.

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2.0 Literature Review

2.1 Isotopic Separation

This research requires the use of an isotopic separation technology. There are two methods of achieving isotopic separation. Isotopes can be separated out of their elemental mass streams using mass differences, or by exploiting electrical or magnetic moments. Mass differentiation has been used for a very long time for the enrichment of uranium. A cost effective method of separating all elemental species would be required for the research in this investigation.

Many candidates for electrical, or magnetic moment, isotopic separation techniques, most of which rely upon selective laser excitation of a chemical or nuclear state, are described in the literature [1]. In one promising technique [2], the material in elemental form is vaporized in a high temperature oven and entrained in a high velocity stream of inert gas. [2].

The jet of air allows for a large amount of target atoms to be entrained; between 5-10% of the initial beam flux. Additional the beam can be pulsed at a rate of 1 kHz, which allows for a near continuous supply of material being passed through the system. The combined stream of inert gas and target material is passed through a 5mm diameter skimmer which is used to focus the beam [1].

Next the high velocity gas is propagated through a laser excitation region where it

is exposed to an array of highly tuned lasers [1]. Each laser is directed through the stream onto a mirror array that redirects the laser back through the stream to increase efficiency. The laser energy is tuned specifically to an energy level for the isotope being separated out. Since the difference in electron energy levels between isotopes is so small, the lasers must have a high degree of precision.

The laser interacts with the electrons of the isotope that is being acted upon to cause it to jump to a higher energy level than ground state. Depending on the atom in question it may undergo another transition or it may stay in the new excited state [1]. The end result of these transitions is atoms of the specific isotope in question are optically pumped to a higher energy state. This change adjusts the magnetic moment of the atoms of the isotope being separated.

The gas stream is then subjected to magnetic field produced by multipole magnets surrounding the separation chamber. These magnetics produce a field that guides the excited atoms toward the outside of the chamber with a force based on properties of the atom.

$$F = \mu_B g_J m_j \nabla B$$

Where μ_B is the Bohr magneton, g_J is the Lande g factor, and m_j is the projection of the total angular moment on the quantization axis [2].

The process has a theoretical enrichment capability of the enriched lithium-6 stream of 95% in a single pass. One pass removed 63.2% of the lithium-6 from the high velocity gas stream. These values are purely theoretical based on a simulation of the process. No experimental data is available. Increased separation efficiencies are obtained by constructing a cascade of separation devices. The level of separation or enrichment depends only on the number of stages the material is passed through and is therefore easily controlled [2].

2.2 Isotopic Transmutation

Previous techniques to burn long lived fission products have not mentioned methods with which to remove the isotopes from their elemental mass streams. As such many investigate the transmutation of species that contain nothing else but the isotope in question. Technetium is a good example of this. The primary isotope of Technetium out of a light water reactor is Tc99 [3], one of the long lived fission products to be removed.

One such paper looks at the transmutation of long lived fission products in a super-critical water-cooled fast reactor [3]. Fission products were selected based on their half-life and their concentration in spent reactor fuel. Of these those that pose a difficulty within the repository were the focus. Theses isotopes were Tc-99, I-129, and Cs-135 due to their ability to transport through rock and ground water[3].

This strategy makes use of blankets loaded with the long lived fission products to be removed. The presence of fission products in the reactor causes the mass balances between the light water reactors and fast reactors to shift. The paper investigates optimum weight fractions for the recycling of fission products. Results show that the technique is low for all isotopes investigated, Tc-99, I-129, and Cs-135. Self-shielding of the isotopes causes the reaction rates to be low; 5.36%, 2.79% and 0.53% respectively [3]. This method of removing the fission products resulted in such low transmutation rates that other methods should be explored.

Since the flux of a fast reactor may not be ideal of transmutation of certain fission products. Increased reaction rates may be obtainable through the use of a thermal neutron flux. A separate study investigates the use of a boiling water reactor to burn the fission products. This reactor also makes use of blankets to burn the fission products [4]. Due to the higher absorption rate at lower energies this technique shows transmutation fractions upwards of 20% for Technetium and 7% for the removal of I-129. The transmutation means the number of reactors in the fleet needs to increase in order to handle the increasing weight fraction of long lived fission products inside the reactor.

2.3 MNCP Linear Accelerator Model

The total removal of all long lived fission products from spent nuclear fuel is

required for this investigation. For two of the isotopes investigated only an external source of neutrons is capable of transmuting the isotopes. The external source modeled for this investigation is an experimental accelerator driven system (XADS) [5].

While an accelerator driven system (ADS) is typically used to power a subcritical reactor system, in this investigation the fuel pins are made strictly of the long lived fission products to be burned. Monte Carlo simulations are used in order to effectively model the burn up of the long lived fission products within the fuel pins are used.

The accelerator requires 20 MWe to operate, using a current of 12mA to operate at 1GeV [5]. Protons are accelerated to the energy and collided with a lead target. MCNPX was used to simulate the collisions between the protons and the lead target to obtain neutron leakage rates out of the target and moderator. This was projected as a surface source used to irradiate a subcritical assembly of nuclear fuel composed of minor actinides and fission products.

This research investigated a method of burning two long lived fission products – Tc-99 and I-129 - inside the subcritical assembly. The research showed transmutation rates within the ADS system of the two fission products investigated to be low; Tc-99 at 0.16% and I-129 at 0.44% per full power year inside the ADS [5].

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3.0 Methodology

These isotopic partitioning strategies are investigated in the context of a closed fuel cycle consisting of a symbiotic fleet of light water reactors (LWRs) and sodium-cooled fast spectrum burner reactors (FRs). In this cycle, transuranics (TRU) are fissioned in the FRs at the same rate as they are discharged from the LWRs.

Chemical separation of TRU and uranium from fission products is an integral part of this reference cycle. For simplicity, this analysis assumes that additional chemical processing to separate the element bearing the isotope can always be achieved. Additionally the reactor fleet is at equilibrium when isotopic separation is implemented.

The simulations are done using the Bright fuel cycle simulator called Bright [1]. Bright has a fluence-based reactor physics capability that computes discharge isotopic vectors for LWRs and FRs for any initial fuel composition inside the domain of a pre-computed library. Using this reactor material balance capability, Bright iteratively arrives at equilibrium reactor support ratios and fuel cycle material flows.

The light water reactor loads 4% enriched fuel with a 3 batch fuel management and a burnup of 51MWd/kgIHM. Fuel from the light water reactor is stored for five years. The fuel then passes to a reprocessing module where specific subspecies from the LWR output isotopic vector are separated to predetermined efficiencies. For this analysis

all species separated are done so with 99% efficiency, except for uranium which is separated with an efficiency of 99.9%. The TRU from the fuel are then passed into a fast reactor.

The input fuel to the fast reactor is a mixture of the TRU from the light water reactor used fuel (UF), self-recycled actinides from discharged fast reactor used fuel, and a depleted uranium top up mass stream. The sodium-cooled fast reactor targets a burnup of 180MWd/kgIHM, although burnup may in practice be limited by fast neutron fluence. Fast reactor fuel is stored for five years prior to reprocessing.

Bright derives the isotopic content of the high level waste mass stream for the equilibrium fleet. Using the output isotopic vectors from the light water reactor and the fast reactor, the mass stream is made up of the fission products from both of the reactors and the losses from the chemical separation processes. It is assumed that the separated uranium is sent to a dedicated storage facility.

REFERENCES

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3.1 ACTINIDE DECAY RECYCLING STRATEGY

This investigation looks at two modifications to the fuel cycle. The first modification (Figure 3.1) involves the partitioning of an intermediate-lived actinide isotope from the fuel cycle. Curium-244 will be used to illustrate the concept and its objectives. This isotope is a major contributor to the specific decay power and neutron radiation field of recycled TRU fuel. When recycled, it is a gateway to higher TRU isotopes – long-lived Cm species through Cm-248 as well as Bk and Cf – that further exacerbate both short-term fuel handling and long-term waste disposability issues.

In the first strategy considered here, the Cm-244 isotope is separated from an elemental Cm stream. The Cm-244 is placed in decay storage, while the balance of the Cm isotopes are returned to the FR and mixed homogeneously with the other recycled TRU. The Cm-244 decay product, Pu-240, is harvested from the Cm-244 decay storage and the Pu-240 is added to the feed stream for the FR fuel. This strategy is feasible because the 18 year half-life of Cm-244 means that secular equilibrium will be approached within several decades. At that time, Pu-240 will be recycled into the FR fuel at the same rate that Cm-244 is partitioned from it. This strategy is advantageous for the fuel cycle because it has the potential to greatly reduce the equilibrium concentration of not only Cm-244 but also its heavier activation products. The strategy will also be investigated for Pu-241, where the decay daughter to be recycled is Am-241.

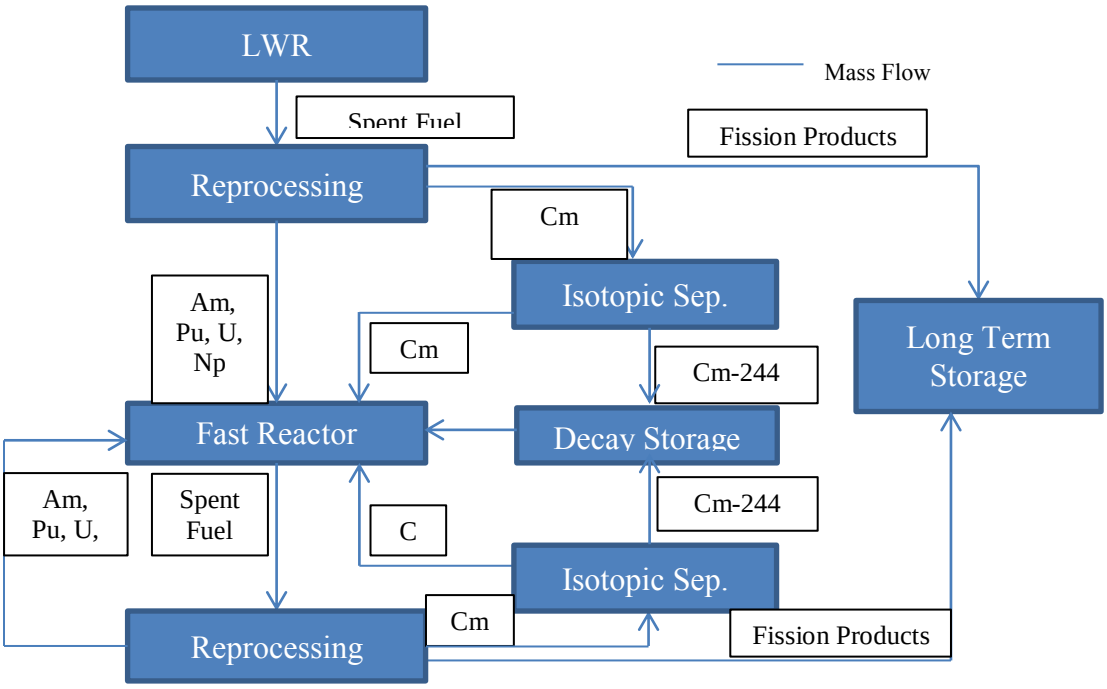

Figure 3.1: Actinide Partitioning and Decay Storage Fuel Cycle

3.2 Long Lived Fission Product Transmutation

The equilibrium phase of the SCFBR fissions transuranic material at the same rate it is being produced in the LWR thus prevent these heavy metals from entering into the high level waste mass stream. The only actinides that end up in the repository are the losses from the chemical separation process; it is assumed that one percent of the material being separated out remains with the waste stream.

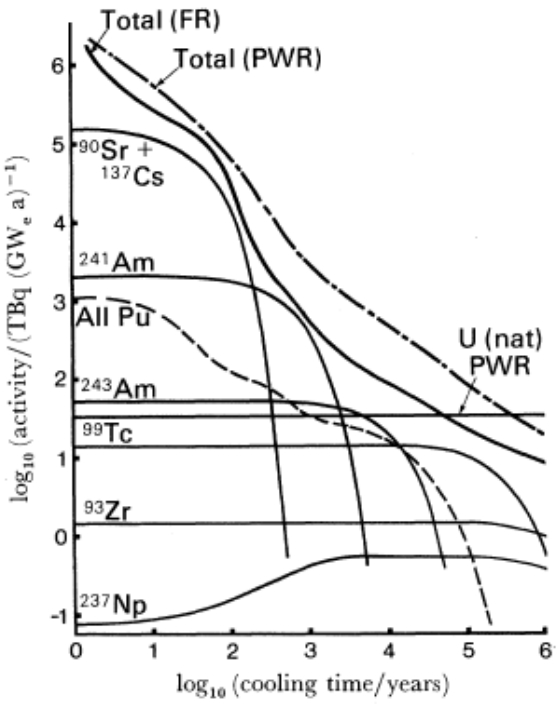

Figure 3.2.1: Radioactivity of select species up to 1 million years [2].

Burning spent fuel in this manner greatly reduces the amount of long lived actinides in nuclear waste. This technique fails to address the fission products created in the LWR. A majority of these fission products are short lived that decay off within a 200 year period. However, there are some longer lived fission products with half-lives comparable to the actinides; these fission products can cause issues with long term geological disposal [1]. These fission products are source of decay heat and radioactivity Additionally should the repository fail, they can be easily transported with the ground water through the surrounding rock layer [1].

Long lived fission products to be investigated were determined using their half-lives and abundance. All fission products of sufficient concentration – mass fraction greater than 1E-6 –from the light water reactor and fast burner reactor spent fuel mass streams were considered as candidates for transmutation. From these only isotopes with a high life of over 100 years were selected.

Rad HLW 10k yr (Ci/kgIHM)	% of total
TC99	46.31
PU240	18.55
PU239	14.72
ZR93/NB93M	12.25
NP239	2.22
AM243	2.22
SE79	1.35
PD107	0.55
PU242	0.33

Table 3.2.1: Radioactivity Faction at 10k years

Two factors weigh heavily on the long term safety of the repository; radioactivity [1] and migration of radionuclides [3]. Table 3.2.1 shows the top 10 contributors to the radioactivity of the high level waste mass stream at 10,000 years. The table was constructed using data for the high level waste material using the base case closed fuel cycle. At these time scales a mother/daughter isotope to come into secular equilibrium with each other; these isotopes are listed together. Separation of the parent isotope will remove the daughter isotope as well.

Table 3.2.1 shows four long lived fission products that contribute to the radioactivity at 10k years: Tc99 (46.31%), Zr93 (12.25%), Se79 (1.35%), and Pd107 (0.55%). Tc99, Cs135 and also I129, are particularly susceptible to chemical migration issues should they leak out of the geological repository [3]. Finally, figure 3.2.2 demonstrates Cs135 and other fission products contribute to the radiotoxicity over the lifetime of a repository [4].

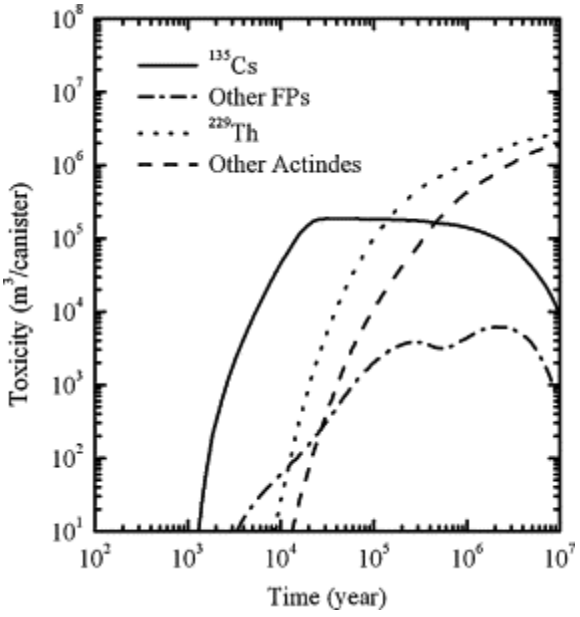

Figure 3.2.2: Radiotoxicity of High Level Waste [4]

3.2.1 Long Lived Fission Product Transmutation in a Fast Reactor

Once the fission products have been partitioned, several options exist to transmute them. A diagram of the basic mass flows of this fuel cycle can be seen in figure 3.2.3. The diagram represents a strategy in which all fission products are recycled together. It is possible to recycle each fission product separately or in different combinations as well.

For this study it is assumed that the addition of fission products to the fuel will not have repercussions on fuel rod strength or integrity. This ensures that no other adjustments will need to be made to the reactor that would have impacts on the neutron economy and the heat transfer characteristics.

The fuel blending strategy requires that the fission products reach equilibrium concentration within the entire fuel cycle. The overall production rate within the light water reactor will remain unchanged under this implementation, however due to the addition of fission products to the fast burner reactor the rate at which fission products are burned and produced in the FR will be noticeably different.

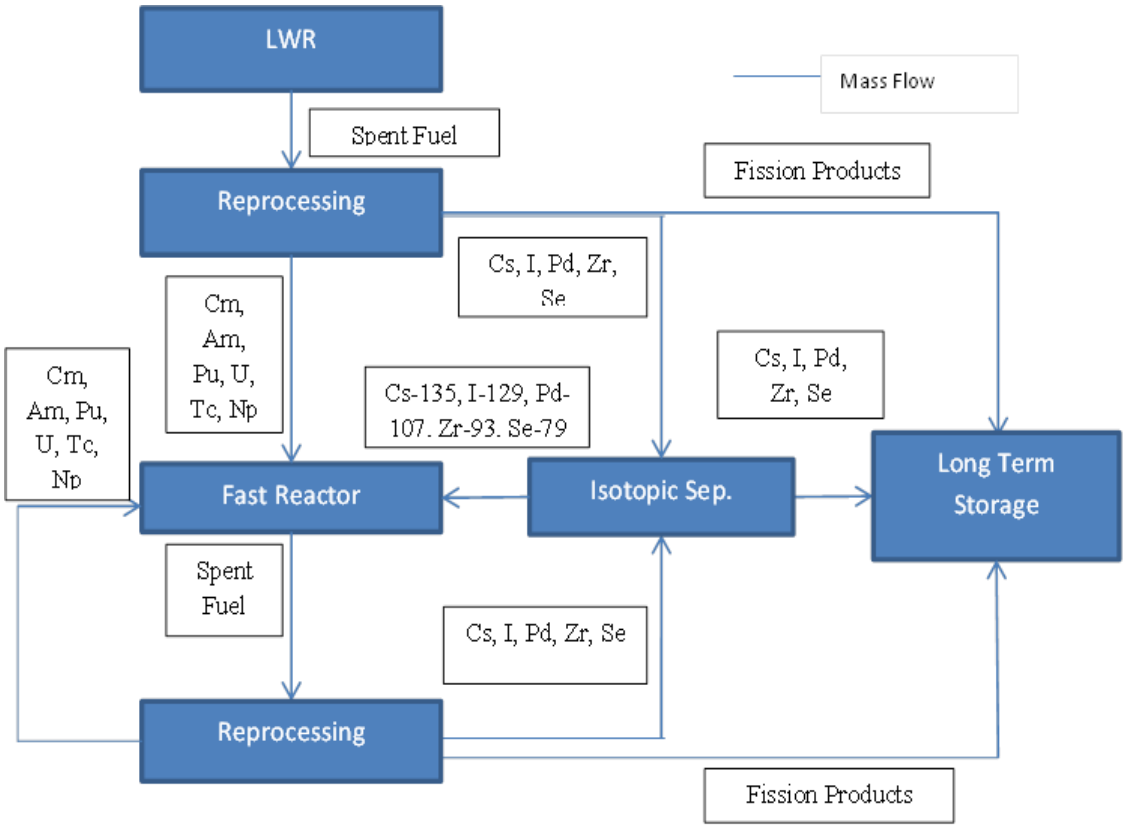

Figure 3.2.3: Long lived fission product burning in fast reactor

Fission products act as neutron poisons in the nuclear fuel. This presents a major problem to the physics of the reactor. Primarily the addition of the fission products will have negative impacts on the neutron economy [4]. Burning fission products robs the reactor of neutrons that could be used to fission the transuranics making it difficult for the reactor to remain critical. Therefore it is important to determine if the fast reactor modeled in this study is capable of supporting the equilibrium concentration of a long lived fission products in the fuel without causing severe changes to the behavior of the reactor.

The rate at which the long lived fission products are being produced or burned in the reactor can be estimated from;

$$\frac{dN}{dt} = \{\sum_{k}(\gamma_{kN}\Sigma_{fkFR}\,\Phi_{FR})\} - (\Sigma_{aNFR}\Phi_{FR}) - \lambda_N N \qquad 3.2.1$$

Where N is the number density of a specific isotope, γ_{kN} is the fraction of fissions of the k[th] fissionable isotope that results in that isotope, Σ_{fk} is the one group fission macroscopic cross section of the k[th] fissionable material, Σ_{aN} is the absorption cross section of the isotope, and Φ is the one group flux of the reactor.

$$\frac{dN}{dt} = \{\sum_{k}(\gamma_{kN}N\sigma_{fkFR}\,\Phi_{FR})\} - (\mathrm{N}\sigma_{aNFR}\Phi_{FR}) - \lambda_N N \qquad 3.2.2$$

$$\frac{dN}{dt} = N * [\{\sum_{k}(\gamma_{kN}\sigma_{fkFR}\,\Phi_{FR})\} - (\sigma_{aNFR}\Phi_{FR}) - \lambda_N] \qquad 3.2.3$$

$$\frac{dN}{N} = dt * [\{\sum_{k}(\gamma_{kN}\sigma_{fkFR}\,\Phi_{FR})\} - (\sigma_{aNFR}\Phi_{FR}) - \lambda_N] \qquad 3.2.4$$

$$N = N_0 e^{t*[\{\sum_k(\gamma_{kN}\sigma_{fkFR}\Phi_{FR})\}-(\sigma_{aNFR}\Phi_{FR})-\lambda_N]} \qquad 3.2.5$$

Since no changes are being made to the light water reactor the rate at which this material is being created is a constant. To account for the fission product being produced in the light water reactor, the amount produced in the LWR is added directly to the fast reactor each time the fast reactor is refueled from the light water reactor spent fuel.

$$N = N_0 e^{t*[\{\sum_k(\chi_{kN}\sigma_{fkFR}\Phi_{FR})\}-(\sigma_{aNFR}\Phi_{FR})-\lambda_N]} + LWRmass \qquad 3.2.6$$

Equation 3.2.6 takes the form of a step function that approaches an equilibrium value. LWRmass in this equation is the amount of the fission product isotope in question that results from the amount of light water reactors that are required to provide the fast reactor with fuel. This value depends on the power share of the two reactor fleets. As the amount of fission product within the fast reactor increases toward equilibrium it will have less room for fuel from the light water reactors. This drives a small feedback effect on the amount of light water reactors required to drive a burner reactor. This effect is ignored for this simple calculation; however it is taken into account during the Bright simulation.

From equation 3.2.6 it is possible to estimate if the equilibrium concentration is too high for a SCFBR to support burning the fission products and still be a critical system. Any fission product with an equilibrium mass fraction of over ten percent of the fuel cannot effectively be burned using this method.

For isotopes that are determined to be suitable for FR transmutation, the Bright code [5] will used to calculate the reactor isotopic output vectors. These vectors will then be passed to the Origen 2.2 code [6] to provide information on the fuel cycle metrics of importance.

3.2.2 Long Term Fission Product Transmutation via Spallation Neutron Source (SNS)

The difficulty of transmuting certain long lived fission products within the sodium cooled fast reactor can be overcome through the use of a spallation neutron source. The spallation neutron source does come with its own complications. The lack of fissionable isotopes means it is not possible to produce energy using the facility. Typically this type of facility would have a subcritical reactor that can be used to produce electricity. The transmutation of fission products does not products electricity. Therefore the operation of such a facility will increase the overall cost of the fuel cycle due to the large amount of energy required to run a linear accelerator spallation neutron source (LINAC SNS). This additional cost will need to be weighed against the benefit to repository costs in order to determine if this is a viable option for eliminating the remainder of the long lived fission product waste.

LINAC SNS facilities will also have an upfront capital cost that must be considered [6]. Most alternative techniques discussed in this paper do not require the construction of new facilities. Thus the number of LINACs required to burn the remaining LLFP must be determined such that their cost can be incorporated into the analysis.

Determining the total number of LINAC SNS required to burn all of the fission products requires knowledge on the rate of transmutation per facility for each isotope. To determine the burn up rates for the LINAC SNS burners it would be important to determine the flux that the facilities are capable of achieving. The rate of change of the

isotope is proportional to the flux that the isotope is subjected to. This relationship is given by:

$$\frac{dN}{dt} = -N\sigma_a\Phi \qquad 3.2.7$$

Where N is the number density of the isotope, σ_a is the microscopic cross section of the isotope and Φ is the flux of the system.

A previous study is referenced to obtain a value for the amount of flux a spallation neutron source can provide in one of these facilities. This study uses Monte Carlo techniques to simulate a spallation neutron source facility [8]. This facility uses a 1 GeV, 12mA proton beam on a natural lead cylindrical target. Outside the target is a graphite moderator with fuel pins located in a hexagon lattice around the target.

Using this study as a basis for the LINAC in this thesis, an MCNP calculation was conducted to simulate the rate at which isotopes would be effectively burned inside the reactor. The calculation simulates a lattice of spent fuel pins composed of pure long lived fission product metal. Reaction rate tallies are constructed to calculate the reaction rate within the entire lattice. The reaction rate from MCNP gives a value of barns per cm^2 per proton.

This unit will be converted into energy per mass burned. The conversion proceeds as follows.

$$Rx\left(\frac{b}{cm^2 * p}\right) * \left(\frac{10^{-24} cm^2}{b}\right) * \left(\frac{6.2x10^{18} p}{s * A}\right) * I\ (A) = RxRate\ \left(\frac{1}{s}\right) \qquad 3.2.8$$

The reaction rate is rate at which the amount of material is reduced. This can be represented using a differential equation.

$$dM = M_0 * RxRate * dt \qquad 3.2.9$$

M_0 is the mass of fission product within the reactor when the MCNP tally was calculated which was reactor startup.

The mass of the fission product isotope being removed is assumed to be roughly constant throughout the burn time in the reactor. By multiplying the mass rate of change by the total power of the linear accelerator the equation is converted into units of mass per MJ. In order for this calculation to hold the concentration of the isotope of LLFP in the reactor needs to remain high.

Similar to typical reactors the most efficient way of accomplishing this would be to continuously cycle new material into the transmutation core of each facility. The drawback of continuous recycling is the high cost that would be associated with it [6]. Each time fuel is removed from the LINAC SNS it will need to be reprocessed to remove the long lived fission product from the isotopes it has transmuted into. Fuel that is removed too often will incur a high reprocessing cost, however, fuel left in the LINAC SNS is burned less effectively the longer it remains. A proper balance between the two is necessary if LINAC SNS facilities are to be used effectively.

The nature of the absorption cross section of each isotope will determine how often these linear accelerator facilities will need to be “refueled”. Small absorption cross sections lead to fewer neutron captures and therefore a slower burn up rate for the material. These isotopes will need to be shuffled and topped off with new fission products less quickly than isotopes with higher absorption cross sections.

The actual process of burning these materials would require some down time to both shuffle the fuel and also to add new material in to be burned. It is likely that this shuffling process will not need to take a long time unless the SNS target needs to be replaced or other facility repairs need to be performed. A quick outage period of approximately one week was applied to the end of each burn phase of the material.

Striking a refueling balance requires insight into the behavior of the isotope’s population as a function of time within the transmutation core. For this investigation each isotope was burned under a constant flux at one month intervals. When the burn up rate reaches 50% of the initial burn up rate determined the fuel would need to be reprocessed, or stored for future reprocessing. Using 50% at the removal point, the burn rate is assumed to be 75% of the initial burn up rate for the time the fuel is in the reactor.

This fuel cycle modification will cause some larger changes to the fuel cycle structure than the other technique for burning long lived fission products. After being reprocessed to their individual elemental mass streams the fission products will be

transported to the isotopic separation facilities. Here the waste products – short lived fission products of the same element – are passed onto a repository. The isotope being burned in the LINAC SNS is burned and then this material is sent back to chemical reprocessing to remove the daughter products. Figure 3.2.4 depicts the fuel cycle for this technique.

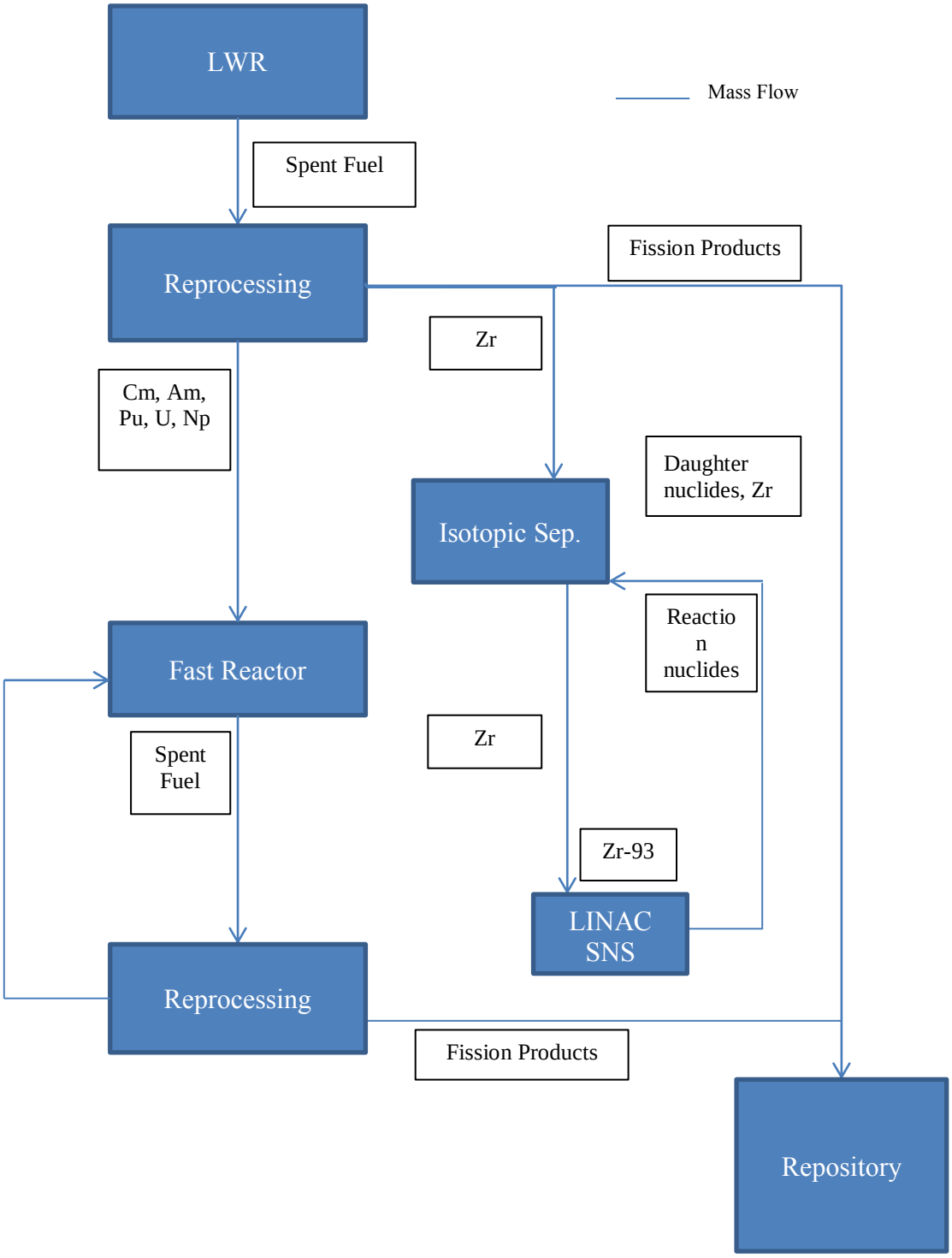

Figure 3.2.4: Long lived fission product removal using LINAC

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3.4 METRICS

To determine the benefit that a change to the reference fuel cycle may cause in the fuel cycle it is important to determine the standard set of metrics with which to compare

the fuel cycles on. These metrics should encompass the main concerns and cost incurring processes of the nuclear fuel cycle. Of particular concern in this study is the back end of the nuclear fuel cycle. Since the LWR input composition does not change in this study the front end metrics of the nuclear fuel cycle will not be considered.

In addition to the back end metrics one intermediate metric will be used to help determine the cost of the fuel cycle. The radioactivity of the material being used to create fast reactor fuel often makes it difficult to make the fuel. Reducing the radioactivity of the material decreases the difficulty of handling and processing new fuel. This is beneficial to cases that recycle fuel, since the fuel being passed to second or third stage reactors will contain radioactive material from the previous reactor stage. Overall radioactivity and spontaneous fission source at the time of fuel fabrication for fast reactor fuel, will serve as metrics for comparing the material at this point in the fuel cycle.

Radioactive material is often a major concern with in the nuclear fuel cycle due to the risks and safety hazards that it can present to society. Reducing the amount of radioactivity in spent fuel is often the goal of many fuel cycle strategies [1]. Due to these risks the level of radioactivity of waste material often has a big influence on the level of safety built into storage facilities. Additional protections built into the facilities to ensure their safety incurs a higher cost. Therefore any reduction in the radioactivity of spent fuel has the benefit of reducing the cost of nuclear power [2].

There are several points of importance during the lifetime of spent fuel in a repository. The first such point is the radioactivity of the fuel when it is taken out of interim storage and ready for transport to the permanent storage facility. Transportation of large quantities of highly radioactive spent fuel can be difficult and costly [2].

After 100 years within the repository the short lived fission products will have decayed out of the spent fuel. In current strategies the approach to dealing with spent fuel is to actively cool it for 100 years. After this 100 year period the nuclear waste should have a low enough radioactivity such that active cooling is no longer required [2]. However, since the material is still radioactive it will continue to emit heat, albeit at a much lower rate. Therefore both the overall radioactive and the spontaneous fission source at 100 years will serve as useful metrics.

Due to the difficulty of understanding if an engineered structure can survive for the 10k years that the repository is required to last, the radioactivity of the material in the repository at that time is important to know. If the radioactivity is reduced to the level of natural uranium at this point failure of the repository is not a concern. At this point the repository is no worse for the environment or society than the original ore from which the uranium is mined. If however, the radioactivity is above the level of natural uranium there are concerns that storing the material is not feasible [2]. This can be applied to the radiotoxicity as well, since removal of the fission products eliminates their source of radiotoxicity.

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3.5 ECONOMICS

The cost of large scale high purity isotopic separation using laser excitation is unknown. The objective of this analysis is to determine the maximum price that can be charged for this process which still results in a reduction in the overall cost of producing nuclear electricity.

The Cm-244 strategy aims to reduce the radiation present at the fabrication of fast reactor fuel, a costly process for remote handled metal fuel [1]. Alleviating this cost will have a large impact on the overall cost of the nuclear fuel cycle.

The Cm-244 strategy increases the amount of Pu-240 being fed into fast reactors. This combination causes a slight change to the reactor power split at equilibrium. Hence it is not sufficient to consider just fuel cycle cost; the total cost of producing electricity must be used.

The LLFP partitioning strategy aims to reduce the cost of the geological repository. At equilibrium the actinides are burned at the same rate at which they are produced, therefore only the waste bring transmuted from reprocessing is sent to storage. If the long lived fission products are also in equilibrium, the burden that they placed on the fuel cycle is removed.

In this partitioning scheme we assume that the long lived actinides are also fully transmuted. The result of this is that the high level waste will be made up of a short lived fission product mass stream. The fission product mass stream can be handled with a less expensive facility than a long term geological repository. Since the life time of this waste is so short the engineering requirements are not as strict, as is the case currently where species with half lives of thousands to millions of years must be isolated from the environment for hundreds of centuries or longer.

Since the waste, after an intermediate period of cooling in surface storage, has such a low level of radioactivity and heat production the design of the facility is less stringent than the long term repository requirements [4].

One additional cost arises to offset this benefit, the cost of the LINAC facilities. Since this technology is not part of the original fuel cycle, the construction and operation costs of the facility will need to be taken into account in the equilibrium electricity cost analysis.

The total cost of running the LINAC will be determined from the amount of energy needed to convert a kilogram of LLFP transmuted. The construction and financing cost of the LINAC will be combined with the operational and fuel fabrication costs. Table 3.5.1 shows the specs of the LINAC used.

LINAC Facility Costs	
Capital Costs	$ 394M
Fuel Fabrication	$ 1000/kg
O&M	$34M/yr
Electrical Demand	20 MWe
Energy	1 GeV
Current	12 mA

Table 4.2.2: Important LINAC Costs

$$LINAC\ COST = Capital\ Cost + Operation\ Cost + Fuel\ Fabrication\ Cost \quad 3.5.1$$

$$LINAC\ COST = \$394M + \frac{\$34M}{year} + \frac{\$1000}{kg}\ [2] \quad 3.5.2$$

The capital cost of the LINAC will be converted into a cost per megajoule using a capital expensive calculation.

$$Capexp = \frac{\left(\frac{Cost}{t} * \sum_{t=1}^{t}(1+IRR)^{t} * \frac{(1+IRR)^{RLT}}{(1+IRR)^{RLT-1}}\right)}{availability} \quad 3.5.3$$

The lifetime of the LINAC is 40 years [2].

$$NAC\ CO \quad = \text{——} + \text{——} \qquad 3.5.4$$

The amount of energy per year used by the LINAC per year is equal to the power of the LINAC multiplied by the total time it is operation. The accelerator operates at 20MWe per year with an availability of 70% [2].

$$\underline{energy} = \qquad \underline{time} \qquad 3.5.5$$

$$\underline{energy} = \qquad \text{——} \quad \text{——} \quad - = \qquad \text{——} \qquad 3.5.6$$

Dividing the annual cost of the LINAC by the annual energy production converts the cost per year to a cost per energy value.

$$NAC\ CO \quad = \left(\text{——} + \text{——} \quad \underline{\text{——}} \right) = \left(\text{——} \right) \qquad 3.5.7$$

The mass of material to be transmuted defines the required LINAC capacity. Therefore the transmutation rate in kg/MJ must be found. This is determined using the MCNPX calculation described previously in section 3.2.2.

Given reaction rates from the MCNP calculations, in MJ/kg of material burned, the LINAC cost component for burning each of the materials can be determined by multiplying this reaction rate by the cost of running the accelerator. Since both isotopes are burned using the LINACs, the total cost is the summation of the cost for burning

each. This can be determined by multiplying the cost per beam energy to burn the material by the operation cost in $/energy. To that the cost of fuel fabrication is added. The end result is the additional cost to the fuel cycle from the LINAC burners.

The rest of the fuel cycle is accounted for using a levelized fuel cycle cost calculation for each equilibrium fuel cycle. Each step in the fuel cycle has a cost per unit measure that passes through the facility. These costs are then summed up and divided by the total amount of kWh(e) produced by the fuel cycle.

The materials may not be used when they are bought. For instance uranium is bought when it is mined. The process to take mined uranium from ore to a fabricated fuel is time consuming. Therefore the return on investment on the ore will not come until the fuel is finally fabricated and burned in the reactor. So levelized life cycle cost analysis will be used to determine fuel cycle and electricity costs.

Therefore the total cost of each fuel cycle can be represented using the following formula.

$$Fuel\ Cycle\ Cost = \frac{\sum_i M_i P_i * (1+\gamma_i)^t}{Electrical\ Power\ Produced} \qquad 3.5.8$$

The costs associated with each of the steps in the fuel cycle are listed in table 1.

Process	unit cost [$]	basis unit
U Mining	30	kgU
Conversion	5	kgU
Enrichment	80	SWU
DU Storage	3.6	kgDU
UOX Fuel Fab	250	kgIHM
SNF Transport	50	kgIHM
SF Reprocessing	1000	kgIHM
VHLW Disposal	2500	kgFP
Metal Fuel Fab	9600	kgIHM
SNF Transport	50	kgIHM
ACT Storage	5000	kgHM
Return on Investment Rate	0.07	-
Waste Disposal Interest Rate	0.025	-

Table 3.5.1: Fuel Cycle Costs for Levelized Fuel Cycle Calculations

These costs are all derived from the Advanced Nuclear Fuel Cycle Cost Basis Report [1]. The costs are scaled to 2009 dollars to avoid the confusion that occurs when dealing with inflation.

The cost of the fuel cycle must be incorporated into the overall cost of electricity for the fuel cycle. The capital costs for the light water reactor and the fast reactor are not included in the fuel cycle cost calculation. Adding capital costs, operation and

maintenance costs, and fuel cycle cost gives the total cost of electricity. Table 3.5.2 gives capital costs for the LWR and FR.

Process	unit cost	basis unit
LWR Capital Cost	3500	$/kWe
LWR O&M	1.8	mills/kWhe
FR Capital Cost	4900	$/kWe
FR O&M	2.0	mills/kWhe

Table 3.5.2: Capital and O&M Costs of Reactors

These values need to be converted to mills ($/1000) per kilowatt hours. Depending on how long it takes to construct the reactor this result can change. The calculation follows as such

$$Capexp = \frac{\left(\frac{Cost}{t} * \sum_{t=1}^{t}(1+IRR)^{t} * \frac{(1+IRR)^{RLT}}{(1+IRR)^{RLT-1}}\right)}{availability} \qquad 3.5.9$$

Where IRR is the interest return rate of capital at 10%, t is the time for construction of the reactor, RLT is the life time of the reactor, and availability is the fraction of time the reactor is operating per year. Using this formula and assuming a construction time of 5 years, availability of 90%, and a reactor lifetime of 40 years the capital costs per kilowatt-hr electric are calculated. Table 3.5.3 shows these capital costs.

Process	unit cost	basis unit
Light Water Reactor	40.98	mills/kWhe
Fast Reactor	57.37	mills/kWhe

Table 3.5.3: Capital Costs of Reactors

Then the cost of electricity is calculated using the following formula.

$$CO = \sum Ca + \sum O + \frac{\Sigma \quad + \gamma}{} \qquad 3.5.10$$

In the first summation the Capexp represents the capital cost of the reactor, and O&Mexp represents the operation and maintenance cost. is the power share of the reactor type in the equilibrium fuel cycle.

To solve for the cost of isotopic separation such that the procedure is economically beneficial to the fuel cycle the cost of electricity of the perturbed fuel cycle is set equal to the reference case cost of electricity.

$$CO = \sum Ca + \sum + \frac{\Sigma \quad + \gamma}{} + \frac{+ \gamma}{} \qquad 3.5.11$$

Pulling the isotopic separation cost out of the fuel cycle cost and solving for price of isotopic separation yields a maximum price that can be changed for isotopic separation.

$$\frac{}{kg} = \frac{}{+\gamma}\left(CO \quad \sum Ca \right) \qquad \sum \quad \frac{\Sigma \qquad +\gamma}{} \Bigg) \tag{3.5.12}$$

Where is the price of isotopic separation for that fuel cycle with units of $/kg input into isotopic separation.

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4.0 Results

4.1 Actinide Recycling Strategies

Table 4.1 shows the top contributors to each of the metrics introduced in 3.4 for the reference fuel cycle. These values were determined by passing the output isotopic vectors of the high level waste mass stream from a Bright fuel cycle to an Origen 2.2 decay deck. The isotopes were then decayed for 100, 1000, 10k, and 1M years. Due to the wide range of the time scales multiple Origen 2.2 runs were required.

It is clear that the primary benefit of partitioning the Cm-244 isotope and recycling its Pu-240 decay product into the fuel cycle would be to mitigate the neutron source at fabrication as well as in the disposed HLW. Note that Cm-244 and its daughters still contribute to the HLW source term since elemental separation efficiencies were taken to be 99% [1]. Therefore, Cm-244 will be chosen as the primary isotope of interest for partitioning and decay, with results for Pu-241 partitioning and decay to Am-241 presented as well. Note that the latter scheme in particular is expected to be detrimental by some measures under this fuel cycle, as at 1% chemical reprocessing loss Am-241 is a strong contributor to intermediate-term disposal metrics.

Rad HLW 100yr (Ci/kWe)	% of total		Rad HLW 1k yr (Ci/kWe)	% of total		Rad HLW 10k yr (Ci/kWe)	% of total
SM151	57.64		AM241	29.43		TC99	46.31
CS137/BA137M	11.68		PU240	23.76		PU240	18.55
AM241	8.78		TC99	23.52		PU239	14.72
PU238	8.68		PU239	8.90		ZR93/NB93M	12.25
Y90/SR90	6.60		ZR93/NB93M	6.07		NP239	2.22
PU240	1.86		NP239	2.55		AM243	2.22
TC99	1.68		AM243	2.55		SE79	1.35
PU239	0.64		SM151	0.79		PD107	0.55

Rad FR Fuel (Ci/kgIHM)	% of total		Spont Fission n source FR Fuel (n/s/kgIHM)	% of total		Spont Fission n source 100yr. (n/s/kWe)	% of total
PU241	70.94		CM244	90.28		CM244	78.43
CM244	16.09		CM246	6.96		CM246	19.20
PU238	9.26		CF250	1.23		CM248	1.69
AM241	1.78		CF252	0.64		PU240	0.32
PU240	0.69		CM248	0.61		PU242	0.22

Table 4.1: Top Contributors to Fuel Cycle Metrics, Reference Scenario

Table 4.2 shows charge and discharge compositions of fast reactor fuel at the equilibrium cycle as computed by Bright. In the table, the first pair of columns covers the reference scheme. The second pair gives the compositions for the Cm-244 partitioning and storage strategy. While there is no Cm-244 in the fresh FR fuel, heavier species (Cm-245 through 248 and higher-Z elements not shown) are still present due to captures in Cm-244 formed during burnup. Because their parent is removed at each recycle, though, the equilibrium prevalence of these heavier species is reduced considerably: approximately 60% for fresh fuel relative to the reference case.

	Reference		Cm 244		Pu-241	
	Weight Fraction		Weight Fraction		Weight Fraction	
	In	Out	In	Out	In	Out
U-235	1.02E-03	8.24E-04	1.07E-03	8.91E-04	1.32E-03	1.11E-03
U-238	6.12E-01	5.35E-01	5.95E-01	5.23E-01	6.45E-01	5.62E-01
Cm-242	6.53E-06	6.47E-06	7.09E-06	7.03E-06	3.27E-06	3.22E-06
Cm-243	4.53E-05	4.05E-05	4.75E-05	4.25E-05	2.62E-05	2.20E-05
Cm-244	6.85E-03	6.37E-03	0.00E+00	2.13E-05	5.05E-03	4.63E-03
Cm-245	1.75E-03	1.73E-03	7.32E-04	7.05E-04	1.31E-03	1.29E-03
Cm-246	6.61E-04	6.57E-04	2.62E-04	2.59E-04	5.62E-04	5.59E-04
Cm-247	4.03E-05	4.03E-05	1.61E-05	1.61E-05	3.54E-05	3.54E-05
Cm-248	1.18E-05	1.18E-05	4.97E-06	4.96E-06	1.17E-05	1.17E-05
Am-241	1.79E-02	1.34E-02	1.91E-02	1.46E-02	1.08E-02	6.90E-03
Am-243	1.35E-02	1.18E-02	1.44E-02	1.27E-02	9.64E-03	8.19E-03
Pu-238	1.86E-02	1.62E-02	2.05E-02	1.79E-02	2.46E-02	2.25E-02
Pu-239	1.40E-01	9.37E-02	1.43E-01	9.52E-02	1.33E-01	9.29E-02
Pu-240	1.04E-01	8.63E-02	1.14E-01	9.49E-02	9.25E-02	7.69E-02
Pu-241	2.37E-02	1.18E-02	2.51E-02	1.27E-02	0.00E+00	7.16E-05
Pu-242	4.03E-02	3.34E-02	4.31E-02	3.60E-02	2.75E-02	2.15E-02
Zr-93	0.00E+00	2.58E-03	0.00E+00	2.60E-03	0.00E+00	2.69E-03
Tc-99	0.00E+00	3.91E-03	0.00E+00	3.94E-03	0.00E+00	3.91E-03
Pd-107	0.00E+00	2.28E-03	0.00E+00	2.30E-03	0.00E+00	2.10E-03
Se-79	0.00E+00	2.62E-05	0.00E+00	2.64E-05	0.00E+00	2.69E-05
I-129	0.00E+00	1.25E-03	0.00E+00	1.25E-03	0.00E+00	1.25E-03
Cs-135	0.00E+00	7.02E-03	0.00E+00	7.07E-03	0.00E+00	7.07E-03

Table 4.2: Isotopic Vectors of Fast Reactor at Equilibrium

Note that the concentration of Pu-240 and its immediate progeny are increased relative to the reference case. Most notably, this gives rise to a 6.7% increase in the concentration of Am-241 in fresh FR fuel. The Bright results indicate that a neutronically equivalent FR fuel under this scheme would load 40.4% TRU, versus

38.7% under the reference scheme. Hence, safety characteristics should be preserved (although this must be verified in subsequent studies) and there is minimal effect on the LWR-to-FR support ratio.

From these material balances, fuel cycle mass balances are obtained and the derived quantities presented for the reference scheme in Table 1 are recomputed. The results for the actinide isotope partitioning and decay strategy are tabulated in Table 3. This table shows the change associated with each metric relative to the reference cycle where no isotopes are partitioned

Fuel Cycle Metric		Reference	Pu241 partitioned	Cm 244 partitioned
Rad FR Fuel (Ci/kgIHM)	total	3.45E+00	9.19E-01	3.07E+00
	% diff	0	-73.3	-11.0
Spontaneous Fission Source (n/s/kgIHM) 0yr	total	7.94E+04	7.30E+04	4.41E+03
	% diff	0	-8.08	-94.4
Rad 100yr (Ci/kWe)	total	5.23E-07	5.99E-07	5.36E-07
	% diff	0	14.5	2.46
Spontaneous Fission Source (n/s/kWe) 100yr	total	5.46E-02	5.75E-02	3.17E-02
	% diff	0	5.20	-4.19
Cumulative Heat (watts/kWe) 1000yr	total	2.28E-02	2.48E-02	2.26E-02
	% diff	0	8.61	-1.07
Rad 10000yr (Ci/kWe)	total	3.33E-08	3.54E-08	3.26E-08
	% diff	0	6.19	-2.05

Table 4.3: Fuel Cycle Metrics, Actinide Isotopic Partitioning Strategies

The removal of Cm-244 offers benefits both before and after and passing through the fast reactor. Table 1 showed that Cm-244 and its activation products provide virtually the entire spontaneous fission neutron source within fast reactor fuel. Therefore, partitioning and decay storage of Cm-244 decreases the radioactivity of fresh fast reactor fuel by 11.0% and spontaneous fission neutron source by 94.4%. These reductions could give rise to significant economic benefits in the fuel fabrication process. The reduction in the spontaneous neutron component of the source term remains considerable even in HLW decayed for 100 years, as the strategy precluded longer-lived neutron-emitting species from being created. Other disposal-related benefits are more modest, as the heavier-than-Cm-244 species were not major contributors to them under the reference scheme.

The Pu-241 partitioning and decay strategy is of less interest under a scheme where the decay product (in this case, Am-241) is recycled homogeneously into the FR fuel since Am-241 is an important contributor to waste-relevant metrics in the 100-1000 year time frame. Indeed, Table 3 shows a worsening of metrics evaluated within this time frame. Pu-241 is the so-called 'gateway' species [2] for formation of all heavier actinides, so its partitioning, along with elemental separation of Am and Cm, would open up multi-tier fuel cycle strategies featuring dedicated MA-burning reactors or subcritical systems in which MA breeding in FRs themselves is minimized.

The reduction in the spontaneous fission source reduces the total cost of the

fabrication of fast reactor fuel. The original cost of the fabrication was taken to be 9600$/kgIHM [3]. This is the high end value assumed for fast reactor fuel fabrication. The assumption made is that the removal of such a large portion of the spontaneous fission source will reduce the cost to the lower end of the projected fuel cycle cost based on the Advanced Fuel Cycle cost basis report[4]. Based on the removal of these fuels the radioactivity of the fabrication material will not be low enough that the use of full remote handling equipment is no longer required. However, a reduction in the amount of shielding required will occur. The reduced price is set to 9000 $/kgIHM. The cost of contact handled fuel fabrication is 7600 $/kg [4]. Since the removal of Cm-244 may not reduce radioactivity enough for contact handled fuel a number close to the high end cost of fuel fabrication was chosen. The cost comparison of this fuel cycle with the standard fuel cycle is recorded in table 4.4.

Fuel Cycle	Cost of electricity (mills/kWhe)	Difference
Standard	55.564	0.00
Cm-244	55.506	-0.11%

Table 4.4: Cost Comparison for Actinide Strategy

Table 4.4a lists the costs and amounts for the fuel cycle components.

	unit cost [$]	basis unit	Amount
U Mining	30	kgU	8.15
Conversion	5	kgU	8.15
Enrichment	80	SWU	5.84
DU Storage	3.6	kgDU	6.9925
UOX Fuel Fab	250	kgIHM	1
SNF Transport	50	kgIHM	1
UOX SF Disposal	500	kgIHM	0
SF Reprocessing	1000	kgIHM	1
VHLW Disposal	2500	kgMA+FP	0.0525
MOX Fuel Fab	9000	kgIHM	0.181
SNF Transport	50	kgIHM	0.181
ACT SF Disposal	5000	kgIHM	.00525
SEP	30500	kgIHM	0.002

Table 4.4a: Mass Balance and Costs Actinide Fuel Cycle

LWR Cap	40.98	Mills/kWhe	0.608
LWR O&M	1.8	Mills/kWhe	0.608
FR Cap	57.37	Mills/kWhe	0.392
FR O&M	2.0	Mills/kWhe	0.392

Table 4.4b Capital Costs of Actinide Fuel Cycle

For this strategy the total cost that can be incurred by using the isotopic separation facility is 0.058 mills/kWhe produced within the whole fuel cycle. This value was arrived at by solving the equation for the price of isotopic separation discussed in section 3.5. The cost of separation from this technique can therefore be determined by increasing the value of separation from zero until the cost of the two fuel cycles is equal. The isotopic

separation cost for the actinide removal strategy that leads to break even cost for the nuclear fuel cycle is 30,500$/kg. Isotopic separation costs below this level would lead to a reduction in cost of the nuclear fuel cycle.

Curium makes up a small portion of the material being feed into the fast reactor. Removing this small portion of the fuel has a large impact on the cost of fast reactor fuel fabrication. That is why the available cost for isotopic separation is so large in this respect.

4.2 Fission Product Recycle Strategies

The second scheme provides a reduction of the long term (ca. 10,000 years and beyond) radioactivity of fission product waste by over 99%, the combined contributions of the isotopes transmuted. This benefit may prove indispensable: regardless of the concept followed for the final HLW repository, it is not feasible to rely upon the integrity of engineered barriers on time scales approaching the duration of climate or geological epochs of the Earth. The only credible pathway for demonstrating the indefinite viability of a repository may thus be to substantially eliminate the source term for 10,000 years and beyond [3].

4.2.1 Fast Reactor Removal

Those isotopes capable of being removed by the fast reactor are assumed to enter

into equilibrium. Therefore the net production of these isotopes will fall to zero. This will leave the fuel cycle with two remaining long lived fission products, Tc-99 and Pd-107, as long lived spent fuel.

4.2.2 LINAC Transmutation

The equilibrium state of the second removal strategy requires the additional cost of constructing the LINAC facilities. Technical specs were described in sections 2.3 and 3.2.2, cost assessment methodology was presented in section 3.5. Table 4.2.2 shows the beam energy that was found to be needed per kilogram of each LLFP to be transmuted.

Isotope	Reaction Rate (MJ/kg)
Tc-99	2025.32
Pd-107	2036.13

Table 4.2.1: LINAC Requirements for Equilibrium

The costs of the LINAC facilities are broken down in table 4.2.2 [5].

LINAC Facility Costs	
Capital Costs	$ 394M
Capital/O&M Cost	$ 0.93/MJ
Fuel Fabrication	$ 1000/kg
Reaction Rate	4061.45MJ/kg
Cost per kilogram	4777.28 $/kg

Table 4.2.2: LINAC Costs

An important note is that if this technology is not paired with the fast reactor

removal strategies the overall price of the fuel cycle will increase due to the cost of building and operating the LINACs. Without the four long lived fission products being removed from the spent fuel there is still a need for a long term repository based on research done for the Yucca Mountain project [3].

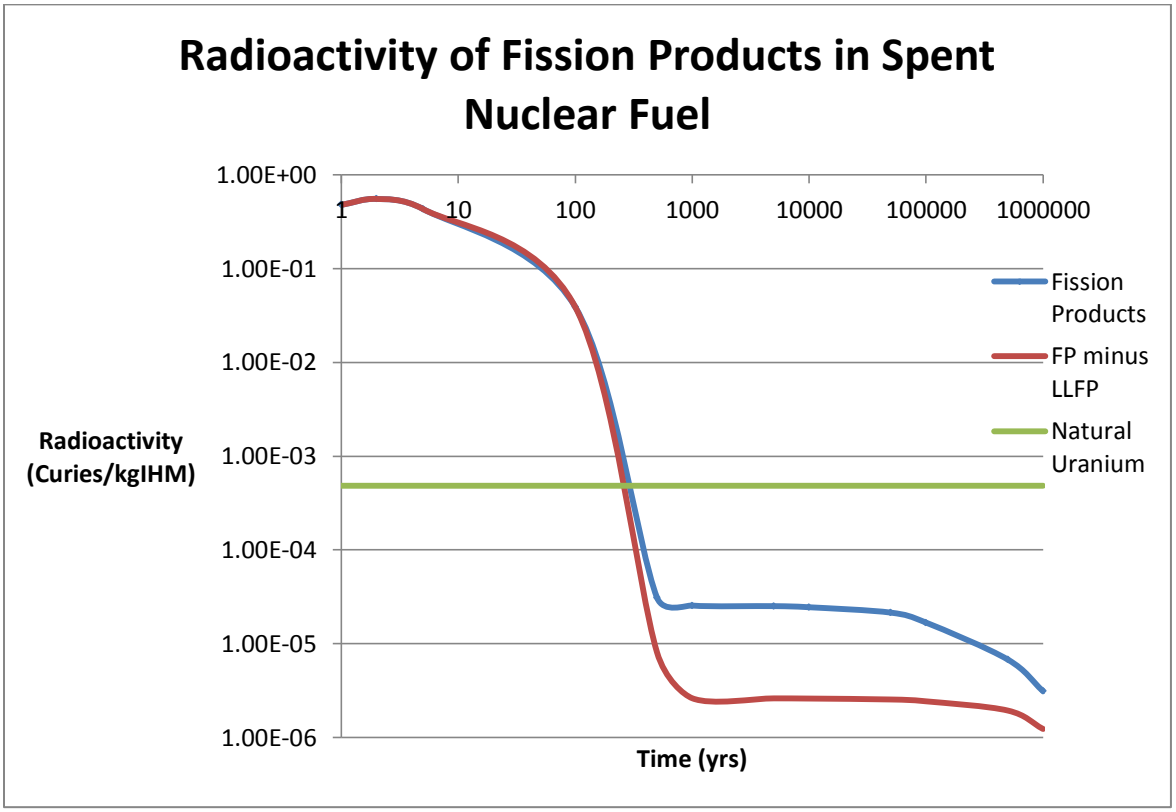

Figure 4.2.1 Fission Product Activity in Repository

The presence of the long lived fission products at time scales pasted 10,000 years was enough to put the feasibility of a long term geological repository into question. Removing the long lived fission products in combination with the actinides removes the need for a facility that can survive millions of years. It is therefore possible to place the

spent fuel in a near surface burial site.

The short lived fission products in the vitrified waste will decay out in approximately 200 years based on Figure 4.2.1 (values calculated using Origen 2.2). At this point only short lived fission products will remain. According to 10CFR 61.55 Waste Classification for waste with Sr-90 with a radioactivity concentration of less than 0.04 curies per cubic meter is capable of being disposed of in near-surface facilities [6]. Sr-90 is the most limiting of fission products by 10CFR 61.55 standards. Since the HLW coming from this fuel cycle is not composed entirely of Sr-90, the value for Sr-90 is accepted as a conservative measure.

A near surface facility is much cheaper than a long term repository buried deep within the earth. Additionally near surface facilities have been built in the past and therefore the technology exists to deal with this type of waste. It is therefore assumed that the waste disposal cost, when incurred following the 200yr interim storage, will be negligible compared to the processing and interim storage costs. See figures 4.2.2 and 4.2.3 for timelines of the reference disposal strategy and FP removal strategy.

The radioactivity of 1 kilogram of fission product material coming from the spent fuel of the reactor at 100 years is 4.543E-5 curies. The density of vitrified waste is 2.65g/ml, or 2650kg/m^3 [6]. Conservatively only 20% of the material in vitrified waste is radioactive. Solving for the amount of curies in one cubic meter;

$$0.20 * 2650 \frac{kg}{m^3} * \left(4.54E-5 \frac{curies}{kg}\right) = 0.0240 \frac{curies}{m^3} \qquad 4.2.1$$

The amount of radiation from fission products at 100 years is less than the 0.04 curies per cubic meter and can therefore be stored in a near surface facility at 200 years. The removal of all mass going to a long term storage facility in this example is justified.

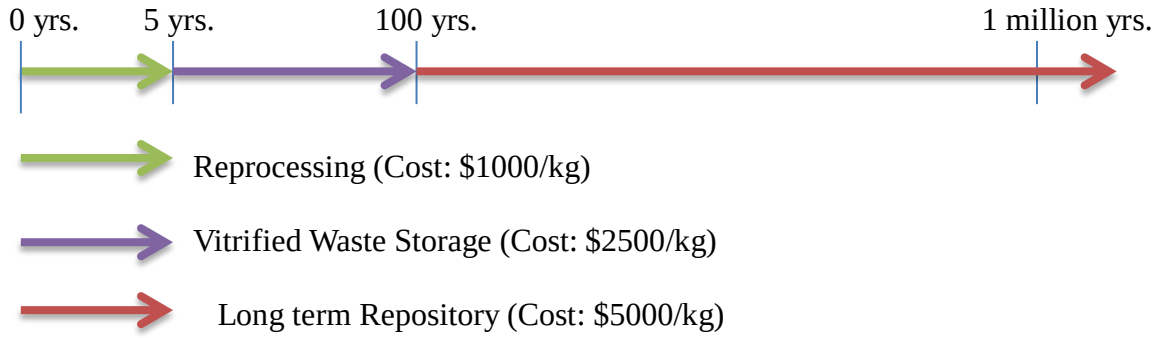

Figure 4.2.2: Waste location through time standard fuel cycle

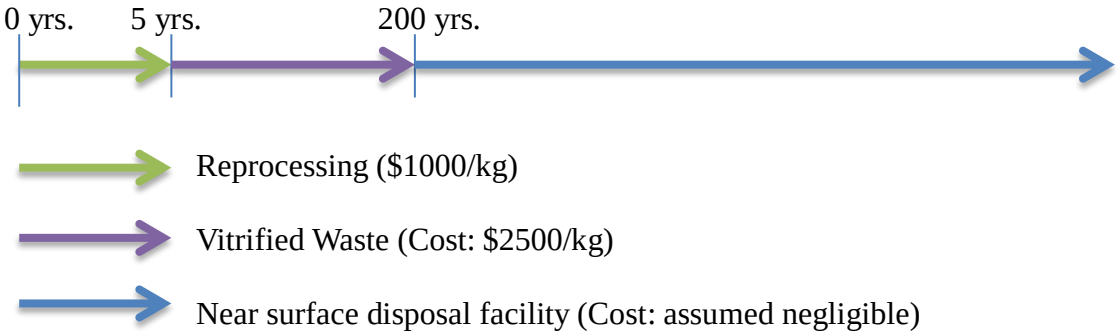

Figure 4.2.3: Waste location through time fission product removal

The LLFP transmutation strategy, paired with full actinide transmutation, turns the issue of radioactive waste into one that can use solutions already in place today. Hence the nuclear fuel cycle no longer needs to rely on a long term repository like Yucca Mountain.

Table 4.2.3 gives the cost reduction for fission products. Table 4.2.3a returns the fuel cycle mass balance costs for the fission product removal strategy.

Fuel Cycle	Cost of electricity (mills/kWhe)	Difference
Standard	59.564	0.00
FP Removal	59.351	-0.38 %

Table 4.2.3: Perturbation Costs

	unit cost [$]	basis unit	Amount
U Mining	30	kgU	8.15
Conversion	5	kgU	8.15
Enrichment	80	SWU	5.84
DU Storage	3.6	kgDU	6.9925
UOX Fuel Fab	250	kgIHM	1
SNF Transport	50	kgIHM	1
UOX SF Disposal	500	kgIHM	0
SF Reprocessing	1000	kgIHM	1
VHLW Disposal	2500	kgMA+FP	0.04
MOX Fuel Fab	9600	kgIHM	0.191
SNF Transport	50	kgIHM	0.191
ACT SF Disposal	5000	kgIHM	0
LINAC	4777	kgIHM	0.0097
SEP	2853	kgIHM	0.012

Table 4.2.3a: Fuel Cycle Mass Balance Costs for Fission Product Removal

LWR Cap	40.98	Mills/kWhe	0.598
LWR O&M	1.8	Mills/kWhe	0.598
FR Cap	57.37	Mills/kWhe	0.402
FR O&M	2.0	Mills/kWhe	0.402

Table 4.2.3b Capital Costs of Fission Product Removal Fuel Cycle

Solving for the price of isotopic separation for this strategy gives a $2,835/kg price for the separation process.

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5.0 Conclusion

This work identified two cases where isotopic separation could offer considerable cost benefits to the nuclear fuel cycle. The first strategy involved the removal of Cm-244 from the fuel cycle to prevent the creation of higher order actinides. The second strategy removed long lived fission products from the nuclear fuel cycle through the use of FR and LINAC transmutation.

The strategy involving the removal of Cm-244 offers great benefit to the nuclear fuel cycle. It reduces the cost of the fast reactor fuel fabrication, which is one of the most expensive components of a closed FR fuel cycle. This strategy showed a maximum price of the separation technology of 30,500 $/kg Cm.

The fission product removal strategy has the benefit of removing the necessity for a long term geological repository as their presence was enough to raise serious doubts about the feasibility of Yucca Mountain. This presents far greater implications than the reduction in price to the fuel cycle. Yucca Mountain has posed a great many research and development issues, in addition to difficult with policy making. These benefits are difficult to quantify, however the cost of isotopic separation for FPs using this strategy was determined to be 2,853 $/kg FP.

Due to the difficult with creating an isotopic separation system the economic benefits to the nuclear fuel cycle that can be gained through the use of this type of technology is rarely considered. This thesis put forth two strategies using isotopic separation and calculated a price that could be associated with the separation technology. Future research could use this method to calculate the benefits and costs of isotopic separation for many other fuel cycles.

One such fuel cycle would be the use of light water reactors to transmute minor actinides. The presence of gateway actinides in light water reactor spent fuel causes difficulties in a thermal flux reactor. By removing these gateway isotopes and allowing those to decay it would be possible to create a minor actinide burning fuel cycle through the use of just light water reactors. By removing the need for expensive fast flux reactors this strategy could provide great benefits to the nuclear fuel cycle.

The small gain to the fuel cycle given by the fission product removal strategy comes due to the ineffectiveness of the LINAC at burning the fission products without an additionally source of neutrons within the reactor. It takes a great deal of energy to burn a single kilogram of material in each LINAC. Therefore a more effective method of burning fission products within a LINAC is needed to make the fission product removal strategy more attractive. One possibility is seeding of the spent fuel with enriched uranium.

There exist additional strategies for the removal of the long lived fission products from the nuclear fuel cycle as well. Homogenous mixing with light water reactor fuel, placement in target locations or blankets in both fast reactors and light water reactors are two examples of additional strategies.

No sensitivity analysis was conducted as part of this investigation. It remains to be seen how the cost of isotopic separation may vary give other perturbations are applied even to the strategies studied in this analysis. Future work would need to include an investigation of the sensitivity to changes and uncertainties in other fuel cycle costs as well as capital costs of the reactors.

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